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RESONANT AND NON-RESONANT OPTICAL FREQUENCY MIXING IN SIMPLE MO--ETC(U)
APR 77 J F WARD AF-AFOSR-2302-72
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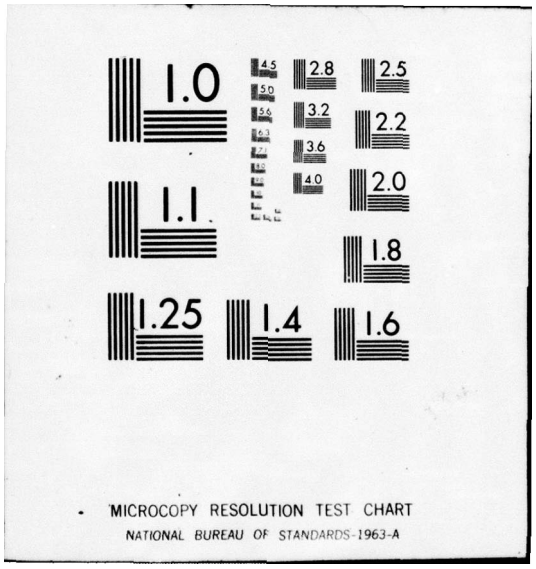
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A technique based on dc-electric-field induced optical second-harmonic generation for measuring second- and third-order electric polarizabilities of molecules in the vapor phase has been developed. The technique has been used to produce detailed and precise data for a wide range of molecules which exist either as gases or liquids at ambient temperature. Hypolarizabilities give rise to phenomena such as optical frequency mixing, modulation and demodulation. Vapors offer technological advantages over crystals for these purposes but the main motivation in our present work is to gain a detailed, fundamental ~over

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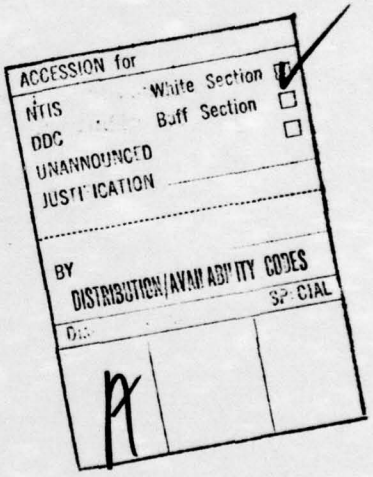
understanding of these properties in the simplest possible molecular systems. This ruby laser radiation is in resonance with two-photon transition in cesium and this system is a useful one in which to study a number of resonant nonlinear optical processes: two-photon absorption and subsequent fluorescence, third harmonic generation, and four-photon parametric oscillation.



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April 28, 1977

Lt. Col. Gordon Wepfer
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AIR FORCE OFFICE OF SCIENTIFIC RESEARCH (AFSC)
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Dear Colonel Wepfer:

Final Report AFOSR 72-2302

Resonant and Non-Resonant Optical Frequency Mixing in Simple Molecular Systems.

I. MOLECULAR HYPERPOLARIZABILITIES (see Journal Articles 1, 2, 3, 5, 10, 11, 12)

We have established and developed a technique based on dc-electric-field induced optical second-harmonic generation for measuring second- and third-order electric polarizabilities of molecules in the vapor phase. The technique has been used to produce detailed and precise data for a wide range of molecules which exist either as gases or liquids at ambient temperature. The molecules studied include:

Inert gases: He, Ne, Ar, Kr, Xe

Thirteen Halogenated methanes: CX_nY_{4-n} n=1-4; X,Y=H, F, Cl and $CBrF_3$

Miscellaneous molecules: H_2, O_2, N_2, CO, NO, H_2S, $(CH_3)_2O$, H_2O, CO_2, NH_3, SF_6.

Hyperpolarizabilities give rise to phenomena such as optical frequency mixing, modulation and demodulation. Vapors offer technological advantages over crystals for these purposes but the main motivation in our present work is to gain a detailed, fundamental understanding of these properties in the simplest possible molecular systems.

For even these simple molecules, ab initio calculations are difficult, typically differing (in the few cases where a calculation has been done) from

experimental results by a factor three whereas the experimental uncertainties are typically better than 10%. Particularly exciting to us is the recent interest shown by Dr. Rod Bartlett of Battelle in improving the ab initio calculations and we look forward to a fruitful interaction here.

We have been interested in less fundamental theories which aim to predict nonlinear properties either from linear properties or from the nonlinear properties of related systems. (This is the only type of approach which has proved useful in predicting the nonlinear optical properties of crystals.) We have been particularly interested in the Bond Additivity Model (based on the notion that a C-Cl bond, for example, has the same nonlinear properties whether it is in a CCl_4 molecule or a CF_3Cl molecule) and in modifications of that model which give useful results without entirely sacrificing simplicity. (See Journal Articles 10, 11, 12.)

Comparison of our vapor-phase measurements with related measurements on liquids is of particular interest since it serves as a probe of the molecular environment in the liquid state.

These topics are all discussed in the Journal Articles. Data to be published in Journal Article 12 is included here (see Appendix).

II. RESONANT NONLINEAR OPTICAL PROCESSES (see Journal Articles 4, 7, 13)

A ruby laser serves as a narrow band source of limited but sufficient tuneability in the region of 6935Å. This radiation is in resonance with the two-photon transition $6^2S_{1/2} - 9^2D_{3/2}$ in cesium and this system is a useful one in which to study a number of resonant nonlinear optical processes: two-photon absorption and subsequent fluorescence, third harmonic generation, and four-photon parametric oscillation.

Resonances offer the possibility of dramatically enhanced nonlinear optical processes. In addition to processes mentioned above, Raman processes, lasing,

optical Stark shifts, multiphoton ionization and distribution of the population over a large number of excited states all occur simultaneously and must be considered together to get a valid picture of the overall interaction. The spatial and temporal dependance of the laser beam and spectral hole burning must also be considered. An understanding of this situation will be fascinating from a fundamental point of view and invaluable technologically in making the best use of resonant enhancement in frequency conversion applications. Details of our present, far-from complete understanding are given in Journal Articles 4 and 7. Work continues on the preparation of Journal Article 13 and Mr. Smith's thesis, which is almost complete.

III. OTHER TOPICS

a) DC-Electric-Field-Induced Optical Second-Harmonic Generation as a Probe of the Focal Region of a Laser Beam (see Journal Articles 6, 8).

Second-harmonic generation in this process depends on a spatial convolution of the dc and optical fields. Since the dc field geometry is known, harmonic generation can be used to yield information on the geometry of the laser beam focus. Air serves as the nonlinear medium and the technique gives detailed information which is difficult to obtain by any other method. Details are given in Journal Articles 6 and 8.

b) DC-Electric-Field-Induced Optical Rectification in Nitrobenzene (see Journal Article 9).

This process results in the generation of a voltage across a Kerr cell during the passage of a laser beam through it. It is theoretically interesting because of its relation to the Kerr effect and, perhaps, of practical interest as a basis for a fast (if insensitive) radiation detector. For details see Journal Article 9.

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5. "Molecular Second- and Third-order Polarizabilities from Measurements of Second-harmonic Generation in Gases," J.F. Ward and Irving J. Bigio, Phys. Rev. A 11, 60-66 (1975).
6. "Electric Field Induced Harmonic Generation as a Probe of the Focal Region of a Laser Beam," Irving J. Bigio, R.S. Finn, and J.F. Ward, Applied Optics 14, 336-342 (1975).
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10. "Measurements of Nonlinear Optical Polarizabilities for Some Halogenated Methanes: The Role of Bond-Bond Interactions," C.K. Miller and J.F. Ward. (Submitted to Physical Review.)

11. "An Interacting Segment Model of Molecular Electric Tensor Properties: Theory and Application to Electric Dipole Moments of the Halogenated Methanes," C.K. Miller, B.J. Orr, and J.F. Ward. (Submitted to J. Chem. Phys.)

12. "Experimental Second- and Third-order Molecular Hyperpolarizabilities," J.F. Ward and C.K. Miller. (Planned for submission to J. Chem. Phys.)

13. "Four-photon Parametric Oscillation in Cesium Vapor," A.V. Smith and J.F. Ward. (Planned.)

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I.J. Bigio, "Studies in dc-Induced Optical Second-Harmonic Generation in Gases," 1973.

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C.K. Miller, "Optical Harmonic Generation in Gases," Research Seminar, U/M, Mar. 1976.

PARTICIPANTS

J.F. Ward	Principal Investigator
K.M. Leung	Graduate Student and Research Associate, 1972. (Now at U.SC.)
I.J. Bigio	Graduate Student 1972 - 1973. (Now at LASL.)
A.V. Smith	Graduate Student, 1972 -
C.K. Miller	Graduate Student, 1973 -
M. Myers	Graduate Student, 1976 -
D. Elliott	Graduate Student, working part time on the project.

The following have also participated on an hourly or unpaid basis:

E. Pellic, D. Kania.

Respectfully submitted,

John Ward

J.F. Ward
Professor of Physics
University of Michigan

sm

APPENDIX

Molecule	$-\Delta k_0$ (cm^{-1})	$10^{36}\ \chi_{\parallel}^{(3)}$ (esu)	$10^{30}\ \chi_{\parallel}^{(2)}$ (esu)
H_2S	9.73±.10	.865±.022	-.043±.009
NH_3	5.40±.05	.512±.010	-.210±.004
$(CH_3)_2O$	8.92±.09	.823±.030	-.468±.011
CO	3.35±.03	.144±.004	.129±.013
NO	2.72±.03	.235±.007	.153±.018
H_2O	2.42±.02	.192±.010	-.094±.003
H_2	1.27±.01	.0652±.0008	--
N_2	1.89±.02	.0866±.0010	--
O_2	2.41±.02	.0953±.0016	--
SF_6	3.47±.03	.130±.002	--
CO_2	3.36±.03	.1119±.0013	--